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## Research article

## Scale up and scale down issues of renewable ammonia plants: Towards modular design

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## ABSTRACT

In this work, the scale-up and down of a renewable-based ammonia facility has been evaluated. Nitrogen is obtained from air separation. Three technologies have been compared, membrane separation, pressure swing adsorption and a Linde's double column. Hydrogen is produced from water splitting using solar, photovoltaic, or wind energy. Finally, ammonia is synthesized in a three bed packed reactor. Two reactor designs were evaluated, direct and indirect cooling. The process is optimized by solving an NLP for each reactor design and nitrogen technology combination and for several production capacities, evaluating the operating and investment costs resulting from the need to use a number of parallel units. The production capacity defines the best technology depending on its characteristics. The results show that for very low production membranes are recommended, medium capacities should be produced using PSA while large require the use of distillation. The actual transition points have been computed. The high costs of panels and electrolyzers mitigate the issues related to duplicating small units. The expected decrease in the cost of both will result in competitive renewable ammonia production costs. Correlations for the investment and production costs as a function of the scale have been developed.

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## 1. Introduction

Renewable sources of energy such as biomass, solar, wind or geothermal, just to mention some of the most widely extended, are characterized by a highly distributed production across regions (EPA, 2017). Total renewable energy available is more than enough to provide for society needs, but the traditional production paradigm is changing. Economies of scale have featured current industry and its infrastructures based on large production complexes (i.e Dow, Exxonmobil or BASF hubs). The well-known six tenths rule has extensively been used in the chemical industry to scale up or down the cost of technologies (Douglas, 1988). However, distributed production does not follow this rule. Distributed production also corresponds to the production at small scales (Pepermans et al., 2005). This new production scheme results in the use of a number of individual units so that the cost is no longer a continuous function. The step forward is modularization of chemical plants. The plants will be built in the form of modulus that are easily assembled at any place to make the most of distributed resources (Baldea et al., 2017). The advantages of these plants are straightforward, easy and quick deployment and low investment risk to exploit resources even in remote places. However, the disadvantages must be also considered including the environmental

impact of the transportation of the plants and of the products (EPA, 2017).

In this context, the production of ammonia is a good example of the use of renewable power to obtain a basic chemical that is used as a raw material for a number of industries, including fertilizers, explosives and plastics. Large ammonia plants based on fossil resources dominate industry (Appl, 2011). Alternatively, nitrogen and hydrogen can be produced from renewable sources (Sánchez and Martín, 2018). The economy of renewable based chemicals are heavily affected by distributed generation. Renewable hydrogen can be obtained from biomass (Martín and Grossmann, 2011) or using solar (Davis and Martín, 2014a; Levene et al., 2005) or wind energy (Davis and Martín, 2014b; Levene et al., 2006) through water splitting. Air separation to produce nitrogen is a mature process too. The technology used to separate air into its components depends on the scale and the purity (Ivanova and Lewis, 2012). Large scale production is typically carried out using Linde's double column. Non cryogenic separations require less capital investment. According to the leaflets of the air separation industry, technologies like pressure swing adsorption or membranes can be used for smaller production capacities, below 500 t/d of oxygen. On the other hand, cryogenic air separation units (ASUs) become cost effective above 200–300 t/d and are more efficient above 500 t/d all the way to 2000 t/d (Matheson, 2018; Messer, 2018), see Fig. 1. However, the purities of the products are jeopardized resulting in losing an important asset, selling oxygen as byproduct. Finally,

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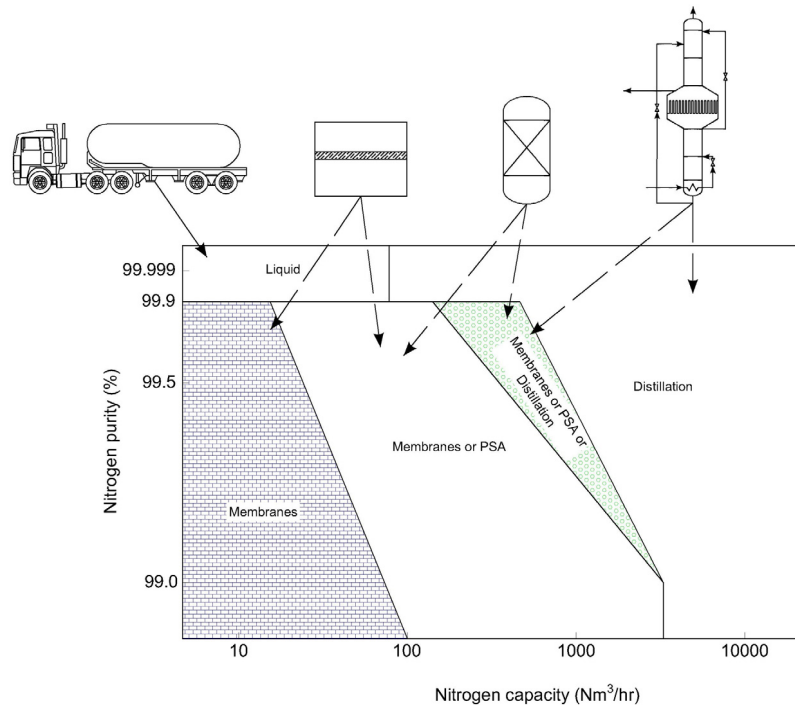


Fig. 1. State of the art of air separation technologies.

ammonia synthesis is carried out in converters characterized by their energy integrated structures since the yield to ammonia is limited by equilibrium.

In this work, a process level analysis for the optimal use of distributed sources of energy at various scales applied to the production of ammonia is performed. The scale-up and down of facilities is evaluated selecting the proper technology for each of the three sections of the plant, evaluating the technologies and their economics. The facility consists of four stages: power collection, considering wind turbines or solar PV panels, air separation for which membranes, PSA or distillation technologies are evaluated, water electrolysis and ammonia synthesis evaluating two reactor designs. The work is organized as follows. Section 2 describes the process flowsheet. Section 3 presents the modeling effort of the membranes and PSA. Section 4 shows the optimization procedure to determine the energy required and the major operating conditions of the units, including the air separation (Distillation, membranes or PSA), the compressors, the ammonia synthesis reactor with its flows, temperature and pressure and the ammonia recovery stage. Section 5 presents some results of the operation, the raw materials and energy requirements, the production and capital cost for scaling-up or down considering modular or non-modular design and a sensitivity analysis on the prices of PV panels or wind turbines. Finally, in Section 6, some conclusions are drawn.

## 2. Ammonia production

The process starts with the technologies that transform solar and wind energy into power. The use of onshore wind turbines or photovoltaic (PV) panels are been considered for this work.

### 2.1. Production of hydrogen

An electrolyzer system is used to split water into hydrogen and oxygen. Two gas streams are produced containing mainly hydrogen and oxygen respectively. Both exit the electrolyzer saturated

with water and with traces of the other species. Most of the water can be removed by condensation. In case of the oxygen stream, after condensation, final dehydration is carried out using an adsorbent bed. Finally, it is compressed for storage. The hydrogen stream is to be further processed to remove the oxygen traces, using a deoxygenation reactor, and final dehydration using zeolites (Davis and Martín, 2014a). Finally, it can be compressed or mixed with the nitrogen.

### 2.2. Production of nitrogen

The production of nitrogen is part of the air separation portfolio of operations in the air separation business. The use of three alternatives is considered: (i) PSA systems for the production of nitrogen, and a byproduct stream rich in oxygen, (ii) membrane separation, producing a permeate and a reject rich in oxygen and (iii) the Linde's double column, suitable for large capacities as presented in previous paper (Sánchez and Martín, 2018). It is assumed that Ar is not recovered.

### 2.3. Ammonia synthesis

The ammonia synthesis stage is widely known as the synthesis loop where the gases are mixed and prepared to be fed to a multibed reactor operating at high pressure. The particular thermodynamics of the ammonia synthesis reaction results in the need to operate in a multibed structure with intercooling (Appl, 1999). Cooling can be achieved either by using cold fresh syngas by means of a more integrated scheme, or by using compressed water producing steam. Thus, direct or indirect reactor designs are considered respectively as a function of the cooling technology. Ammonia is typically recovered by condensation and the unreacted gases must be recovered due to the high production costs of nitrogen and hydrogen. Fig. 2 shows a general scheme of the process.

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